

# Emission of gamma rays shifted from resonant absorption by electron-nuclear double transitions in $^{151}\text{Eu}^{2+}:\text{CaF}_2$

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## Abstract

We show that the emission of a gamma-ray photon by a nucleus can be influenced by a microwave magnetic field acting on the atomic electrons. We study theoretically these electron-nuclear double transitions (ENDTs) for  $^{151}\text{Eu}$  nuclei in a  $\text{CaF}_2$  lattice at low temperature, in the presence of a static magnetic field and of a microwave magnetic field. The ENDTs acquire a significant intensity for certain resonance frequencies. The ENDTs are of interest for the identification of the position of the lines in complex Mössbauer spectra.

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The possibility of influencing gamma-ray emission with applied oscillatory electromagnetic fields was first analyzed by Hack and Hamermesh. [1] In the present work we show that the emission of a gamma-ray photon by a nucleus can be influenced by a microwave magnetic field acting on the atomic electrons. We consider a two-photon process in which a microwave photon is absorbed by the electrons simultaneously with the emission of a gamma-ray photon by the nucleus. We study theoretically these electron-nuclear double transitions (ENDTs) for  $^{151}\text{Eu}$  nuclei in a  $\text{CaF}_2$  lattice at low temperature, in the presence of a static magnetic field and under the action of a microwave magnetic field. The  $^{151}\text{Eu}$  nucleus is supposed to be initially in the 21.54 keV excited state and the atomic electrons are in the ground state. We show that, under the action of the microwave magnetic field, the coupled electron-nuclear system can make a transition from the ground atomic electron Zeeman level to a higher electron level while at the same time the nucleus makes a transition from the 21.54 keV state to the ground nuclear state emitting a gamma-ray photon. We show that the energy of the gamma-ray photon emitted by ENDTs is shifted away from the parent gamma-ray line, and obtain for the ENDT lines a quadratic dependence on the microwave magnetic field and an inversely quadratic dependence on the detuning between the position of the virtual and intermediate states. The ENDT lines acquire a significant intensity for certain resonance frequencies. The ENDTs are of interest for the identification of the position of the lines in complex Mössbauer spectra.

We shall analyze in this work the 21.54 keV, M1 transition of  $^{151}\text{Eu}$  nuclei embedded in a  $\text{CaF}_2$  lattice. The parity and the nuclear magnetic moment of the ground state are  $5/2+$ ,  $3.474 \mu_N$ , and the spin, the parity, the nuclear magnetic moment and the half-life of the 21.54 keV state are respectively  $7/2+$ ,  $2.591 \mu_N$  and 9.4 ns, where the nuclear magneton is  $\mu_N = 5.05 \cdot 10^{-27} \text{ JT}^{-1}$ . [2] The 18 hyperfine components of the 21.54 keV transition give rise, by overlapping, to an 8-line Mössbauer spectrum.[3],[4] Europium appears in a  $\text{CaF}_2$  lattice as the divalent ion  $\text{Eu}^{2+}$ , having the ground state  $^8S_{7/2}$ . The electron energy levels of the  $\text{Eu}^{2+}$  ion in the cubic field of the  $\text{CaF}_2$  lattice have been studied by Lacroix [5] and experimentally observed with the method of electron paramagnetic resonance by Ryter [6] and Baker et al. [7] The results of ENDOR observations on  $^{151}\text{Eu}^{2+}$  in a  $\text{CaF}_2$  lattice have been reported by Baker and Williams. [8]

We shall write the Hamiltonian of the  $^{151}\text{Eu}^{2+}$  system in the  $\text{CaF}_2$  lattice as the sum  $H = H_e + H_n + H_{en} + V_{mw}(t) + V_\gamma(t)$ , where  $H_e$  is the electron Hamiltonian,  $H_n$  is the nuclear Hamil-

tonian,  $H_{en}$  describes the hyperfine interaction,  $V_{mw}(t)$  describes the interaction of the microwave field with the atomic electrons and  $V_\gamma(t)$  describes the interaction of the gamma rays with the nucleus.

We shall assume that the x, y, z axes are oriented along the four-fold axes of the  $\text{CaF}_2$  crystal, with the origin on a  $\text{Eu}^{2+}$  ion. If the static magnetic field  $B$  is applied along the z-axis, and if we denote the projections of the electron angular momentum along the z-axis by  $M$ , where  $M = \pm 7/2, \pm 5/2, \pm 3/2, \pm 1/2$ , it can be shown [5],[9] that the eigenvalues  $E_M$  of the electron Hamiltonian  $H_e$  are

$$E_{\pm 7/2} = \pm \frac{3}{2} g \mu_B B + 8b_4 - 2b_6 \pm \left( (\pm 2g\mu_B B - b_4 + 3b_6)^2 + 35(b_4 - 3b_6)^2 \right)^{1/2}, \quad (1)$$

$$E_{\pm 5/2} = \pm \frac{1}{2} g \mu_B B - 8b_4 + 2b_6 \pm \left( (\pm 2g\mu_B B - 5b_4 - 7b_6)^2 + 3(5b_4 + 7b_6)^2 \right)^{1/2}, \quad (2)$$

$$E_{\pm 3/2} = \mp \frac{1}{2} g \mu_B B - 8b_4 + 2b_6 \pm \left( (\pm 2g\mu_B B + 5b_4 + 7b_6)^2 + 3(5b_4 + 7b_6)^2 \right)^{1/2}, \quad (3)$$

$$E_{\pm 1/2} = \mp \frac{3}{2} g \mu_B B + 8b_4 - 2b_6 \pm \left( (\pm 2g\mu_B B + b_4 - 3b_6)^2 + 35(b_4 - 3b_6)^2 \right)^{1/2}, \quad (4)$$

where the Bohr magneton is  $\mu_B = 9.274 \cdot 10^{-24} \text{ JT}^{-1}$ , and  $g=1.9926$ ,  $b_4=-176.12 \text{ MHz}$ ,  $b_6=0.78 \text{ MHz}$ . [8],[10] Then the eigenvalues of the time-independent part  $H_e + H_n + H_{en}$  of the Hamiltonian  $H$  are [5]

$$E_{Mm_l}^{(l)} = E_M + A_l M m_l + \frac{A_l^2 (7/2 + M) (9/2 - M) (5/2 - m_l) (7/2 + m_l)}{4(E_M - E_{M-1})} - \frac{A_l^2 (7/2 - M) (9/2 + M) (5/2 + m_l) (7/2 - m_l)}{4(E_{M+1} - E_M)} - g_l \mu_B B m_l, \quad (5)$$

$$E_{Mm_u}^{(u)} = E_0 + E_M + A_u M m_u + \frac{A_u^2 (7/2 + M) (9/2 - M) (7/2 - m_u) (9/2 + m_u)}{4(E_M - E_{M-1})} - \frac{A_u^2 (7/2 - M) (9/2 + M) (7/2 + m_u) (9/2 - m_u)}{4(E_{M+1} - E_M)} - g_u \mu_B B m_u, \quad (6)$$

In eqs. (5),(6),  $E_0=21.54 \text{ keV}$  is the energy of the unsplit excited nuclear state,  $m_l = \pm 5/2, \pm 3/2, \pm 1/2$  are the projections on the z-axis of the angular momentum of the ground state  $l$  of the nucleus, and  $m_u = \pm 7/2, \pm 5/2, \pm 3/2, \pm 1/2$  are the projections on the z-axis of the angular momentum of the excited state  $u$  of the nucleus. The ground state constants are [8],[10]  $A_l/(2\pi\hbar)=-102.9069 \text{ MHz}$ ,  $g_l=7.4968 \cdot 10^{-4}$ . The constants for the upper state  $u$  can be calculated from the relation  $A_u = \mu_u I_l A_l / (\mu_l I_u)$ ,  $g_u = \mu_u I_l g_l / (\mu_l I_u)$ , where  $\mu_l, \mu_u$  are respectively the magnetic moments for

the ground and excited states  $l, u, I_l = 5/2, I_u = 7/2$ , and  $\mu_u I_l / (\mu_l I_u) = 0.5327$ , so that  $A_u / (2\pi\hbar) = -54.8185$  MHz,  $g_u = 3.9936 \cdot 10^{-4}$ . The electron-nuclear states of interest for the present work are represented in fig. 1.

We are now in a position to analyze the two-photon process by which a gamma-ray photon is emitted by the nucleus simultaneously with the absorption of a microwave photon by the atomic electrons. As already discussed, the states of the problem are  $|M, lm_l\rangle$  and  $|M, um_u\rangle$ . In the initial state  $|-7/2, um_u\rangle$  of the two-photon process the  $\text{Eu}^{2+}$  ion is in the  $M=-7/2$  state and the  $^{151}\text{Eu}$  nucleus is in the excited state  $u$  of angular momentum  $m_u$ , and in the final state  $|-5/2, lm_l\rangle$  of the two-photon process the  $\text{Eu}^{2+}$  ion is in the  $M=-5/2$  state and the  $^{151}\text{Eu}$  nucleus reaches the ground state  $l$  of angular momentum  $m_l$ , where  $m_u - m_l = 0, \pm 1$ . The intermediate states of the two-photon process are  $|-5/2, um_u\rangle$  and  $|-7/2, lm_l\rangle$ . We can calculate the ENDT transition amplitude  $c_2 = \langle -5/2, lm_l | \text{ENDT} | -7/2, um_u \rangle$  with the aid of conventional second-order perturbation theory, and the single-photon gamma-ray transition amplitude as  $c_1 = \langle -7/2, lm_l | V_\gamma | -7/2, um_u \rangle$ , where the operator  $V_\gamma$  describes the interaction of the gamma-ray photon with the nucleus. In this way it can be shown that the relative intensity of the ENDT lines  $|c_2|^2 / |c_1|^2$  increases quadratically with the applied microwave magnetic field  $B_{mw}$ , and decreases with the square of the detuning between the virtual states and the corresponding intermediate states. It turns out that significant intensities for the ENDT lines can be obtained when the energy of the microwave photon is resonant with the energy of the transition  $|-7/2, um_u\rangle \rightarrow |-5/2, um_u\rangle$ , so that  $\hbar\omega_{mw} = E_{-5/2, m_u}^{(u)} - E_{-7/2, m_u}^{(u)}$ . The gamma-ray photons emitted in the ENDT transition have the energy  $\hbar\omega_\gamma = E_{-5/2, m_u}^{(u)} - E_{-5/2, m_l}^{(l)}$ , and the positions of the single-photon gamma-ray lines are given by  $\hbar\omega_\gamma^{(0)} = E_{-7/2, m_u}^{(u)} - E_{-7/2, m_l}^{(l)}$ ,

It can be shown by conventional second-order perturbation theory that the ratio  $R = |c_2^{(r)}|^2 / |c_1|^2$  of the square of the resonant ENDT amplitude  $c_2^{(r)} = \langle -5/2, lm_l | \text{ENDT}_r | -7/2, um_u \rangle$  to the square of the unperturbed single-photon amplitude  $c_1 = \langle -7/2, lm_l | V_\gamma | -7/2, um_u \rangle$  is

$$R = \frac{7g^2\mu_B^2 B_{mw}^2}{4\hbar^2 \Gamma^2}, \quad (7)$$

being independent of  $m_u, m_l$ . We consider that the width  $\Gamma$  is determined by the half-life  $t_{1/2} = 9.4$  ns of the 21.54 keV state of the  $^{151}\text{Eu}$  nucleus as  $\Gamma = \ln 2 / t_{1/2}$ . For a resonant microwave field of amplitude  $B_{mw} = 10^{-4}$  T, the relative intensity of the ENDT line is  $R = 0.098$ . The relative variation of the recoil-free fraction for the gamma-ray transitions between the substates shown in

fig. 1 is of the order of  $3E_0\hbar\omega_{mw}/(2m_{\text{Eu}}k\theta_D)$  and is negligible, where  $m_{\text{Eu}}$  is the mass of the europium nucleus,  $\theta_D$  is the Debye temperature of the lattice and  $k$  the Boltzmann constant. In the derivation of eq. (7) it has been implicitly assumed that the half-life of the nuclear excited state is much shorter than the spin-lattice relaxation time, an assumption which is valid for the 9.4 ns half-life of the 21.54 keV state of  $^{151}\text{Eu}$ . [11]

In fig. 2(a) we have represented the ENDT spectrum for a static magnetic field  $B=2$  T, which gives  $(E_{-7/2} - E_{-5/2})/(2\pi\hbar)=59.291$  GHz, for a sample temperature  $T=1$  K. The resonance microwave frequency for the ENDT  $|-7/2, u7/2\rangle \rightarrow |-5/2, u7/2\rangle \rightarrow |-5/2, l5/2\rangle$  shown in fig. 1 is  $\omega_{mw}/(2\pi) = 59.098$  GHz. The ENDT line is situated at  $(E_\gamma - E_0)/(2\pi\hbar)=-148$  MHz. The direction of observation is perpendicular to the static magnetic field  $B$ . Assuming a Boltzmann distribution for the electron population, the fractional electron population is 0.941 in the  $M=-7/2$  state and 0.054 in the  $-5/2$  state. The FWHM of the lines was taken 23.5 MHz. The intensity  $|c_1|^2$  of the single-photon lines is proportional to  $|(\mu_+)_{lm_l, um_u}|^2 + |(\mu_-)_{lm_l, um_u}|^2 + |(\mu_z)_{lm_l, um_u}|^2$ . In fig. 2(b) we have represented the ENDT spectrum for a static magnetic field  $B=0.2342$  T, which gives  $(E_{-5/2} - E_{-7/2})/(2\pi\hbar)=10$  GHz, and at a sample temperature  $T=1$  K, for the same ENDT transition and direction of observation as before. The resonance frequency is 9.797 GHz, and the ENDT line is situated at  $(E_\gamma - E_0)/(2\pi\hbar)=-142$  MHz. The fractional electron population is 0.31 in the  $M=-7/2$  state and 0.19 in the  $M=-5/2$  state. The relative intensity of the ENDT line, being proportional to the difference of the electron populations of the  $M=-7/2$  and  $M=-5/2$  levels, is thus smaller at 10 GHz than at 59 GHz, but the ENDT contribution can still be clearly seen in the spectrum.

The process by which the  $^{151}\text{Eu}^{2+}$  system, initially in the  $|-7/2, u7/2\rangle$  state, absorbs a microwave photon up to the  $|-5/2, u7/2\rangle$  intermediate state before emitting a gamma-ray photon to reach the final  $|-5/2, l5/2\rangle$  state is labelled in fig. 1 as direct ENDT. The cross section for the stimulated emission of a gamma-ray photon via this ENDT is denoted by  $\sigma$ . There is also an ENDT when the  $^{151}\text{Eu}^{2+}$  system, initially in the  $|-5/2, l5/2\rangle$  state, absorbs a gamma-ray photon up to the intermediate  $|-5/2, u7/2\rangle$  state and then emits a microwave photon to reach the final  $|-7/2, u7/2\rangle$  state. The process is displayed in fig. 1 as the inverse ENDT. The cross section for the absorption of a gamma-ray photon via the inverse ENDT will have the same value  $\sigma$  as the direct ENDT.

Absorption ENDTs are also possible when the  $^{151}\text{Eu}^{2+}$  system, initially in the  $|-7/2, lm_l\rangle$  state, absorbs a microwave photon up to the intermediate state  $|-5/2, lm_l\rangle$ , and then absorbs a gamma-ray photon to reach the final  $|-5/2, um_u\rangle$  state. For a resonant ENDT in absorption, the energy of the absorbed gamma-ray photon continues to be given by  $\hbar\omega_\gamma = E_{-5/2, m_u}^{(u)} - E_{-5/2, m_l}^{(l)}$ , and the relative intensity of the absorbed gamma ray by eq. (7), but the microwave resonance frequency is, for a direct ENDT in absorption,  $\hbar\omega_{mw}^{(a)} = E_{-5/2, m_l}^{(l)} - E_{-7/2, m_l}^{(l)}$ . The resonance microwave frequency for the direct ENDT in absorption  $|-7/2, l5/2\rangle \rightarrow |-5/2, l5/2\rangle \rightarrow |-5/2, u7/2\rangle$  is  $\hbar\omega_{mw}^{(a)}/(2\pi)=59.032$  GHz for  $B=2$  T, and  $\hbar\omega_{mw}^{(a)}/(2\pi)=9.717$  GHz for  $B=0.2342$  T. The difference  $\delta = (\omega_{mw} - \omega_{mw}^{(a)})/(2\pi)$  is  $\delta=66$  MHz for  $B=2$  T, and  $\delta=79$  MHz for  $B=0.2342$  T.

The ENDTs described in this work may have applications for the research on the amplification of gamma rays without inversion of nuclear population. [12] Another interesting application of ENDTs would be the observation of gamma-ray *lines* in situations when the conventional Mössbauer spectrum is unresolved, or partially resolved. This case is exemplified by the spectrum of fig. 2(b), where there are many overlapping lines arising from the electron states of various  $M$  values. If we record Mössbauer spectra for a series of applied microwave frequencies from a range where we expect to have resonances at the value of the applied static magnetic field, the difference between these spectra and a reference spectrum corresponding to a non-resonant microwave frequency will show the ENDT contribution, as illustrated in the lower part of fig. 2(b). For a particular resonance frequency, this contribution consists of pairs of peaks and dips, the positions of which coincide with the position of the lines in the  $M=-7/2$  and respectively  $M=-5/2$  Mössbauer spectra originating from *one* initial nuclear sublevel. Therefore, the ENDT peak/dip spectrum is much simpler than the full Mössbauer spectrum, where we have lines from *all* values of  $M$ . Thus, in the ENDT approach, the full Mössbauer spectrum is decomposed in a series of partial Mössbauer spectra obtained for the microwave resonance frequencies of the system.

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## FIGURE CAPTIONS

Fig. 1. Energy levels of the electron-nuclear system  $^{151}\text{Eu}^{2+}:\text{CaF}_2$  relevant for the present work. For a static magnetic field  $B=2$  T we have  $(E_{-5/2} - E_{-7/2})/(2\pi\hbar)=59.291$  GHz,  $\omega_{mw}/(2\pi)=59.098$  GHz,  $\omega_{mw}^{(a)}/(2\pi)=59.032$  GHz. For a static magnetic field  $B=0.2342$  T we have  $(E_{-5/2} - E_{-7/2})/(2\pi\hbar)=10$  GHz,  $\omega_{mw}/(2\pi)=9.797$  GHz,  $\omega_{mw}^{(a)}/(2\pi)=9.717$  GHz.  $E_0=21.54$  keV is the energy of the excited nuclear state of  $^{151}\text{Eu}$ .

Fig. 2. Unperturbed Mössbauer spectrum of  $^{151}\text{Eu}^{2+}:\text{CaF}_2$  (upper curve) and ENDT contribution (lower curve) at  $T=1$  K, for (a)  $B=2$  T,  $B_{mw} = 10^{-4}$  T,  $\omega_{mw}/(2\pi)=59.098$  GHz, and (b)  $B=0.2342$  T,  $B_{mw} = 10^{-4}$  T,  $\omega_{mw}/(2\pi)=10$  GHz. The FWHM of the lines was taken 23.5 MHz.







